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Original Article

STUDY OF NONLINEAR OPTICAL PROPERTIES OF

NEMATIC LIQUID CRYSTAL MATERIALS

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ABSTRACT

The nonlinear optical properties of nematic liquid crystal material have been studied at different concentrations using Z-Scan technique.Experiments are performed using continuous wave (CW) diode solid state laser at 473 nm wavelength and 20 mW power. In this work, six concentrations were prepared for (Di-Cinnamylidene Benzidine), these concentrations were $(9\times10^{-5}, 8\times10^{-5}, 7\times10^{-5}, 6\times10^{-5}, 5\times10^{-5})$ and 4×10^{-5})M. The optical absorption and transmission spectra for these concentrations were measured by using UV-VIS spectrophotometer. Our results show that the nonlinearity are changed significantly with concentrations, and the capability using the sample as optical limiter device.

KEYWORDS: Nematic Liquid Crystal, Z-Scan Technique, Nonlinear Refractive Index, Optical Limiting

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INTRODUCTION

Liquid crystals exhibit large photo induced nonlinearity due to their large dielectric anisotropy coupled with the collective director reorientation, Moreover, their optical properties are sensitive to the presence of a small electric or magnetic field. Non-display applications of LCs in optical communication, nonlinear optics, data/signal/image processing and optical sensing are also receiving increased attention[1].

Due to their unique crystalline phase characterized by the partial order of their constituent molecules along with physical fluidity, LCs can be easily incorporated into desired configurations for a variety of device applications, it can be promising materials for optical switching and optical power limiting applications [2]. In the nematic phase the rod shaped molecules are oriented on the average along an axis called the director. The nematic phase has long range orientational order [3]. M.Sheik-Bahae et al. reported the Z-Scan technique for measuring both the nonlinear refractive index and nonlinear absorption coefficient for a wide varity of materials. The simplicity of both the experimental set-up and the data analysis has allowed the technique to become widely used by research groups [4].

The aim of this work was the use of the Z-Scan technique to study the nonlinear optical properties of (Di-Cinnamylidene Benzidine) at different concentrations with (CW) diode solid state laser at 473 nm wavelength and 20 mW power. Also preparing a good limiter device for many applications.

THEORY

The mathematical relationships for nonlinear materials at high intensity of absorption and nonlinear refraction (NLR) is given by [5,6]:

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$$\alpha = \alpha_0 + \beta I \tag{1}$$

Where I is the incident intensity, α_o is the linear absorption coefficient and β is the nonlinear absorption coefficient related to the intensity. At high intensity, the refractive index is given by:

$$n = n_0 + n_2 I \tag{2}$$

Where n_o is the linear refractive index, and n_2 is the nonlinear refractive coefficient. The nonlinear optical properties can be investigated by Z-Scan technique at which it can be used to determine the nonlinear refractive index when closed-aperture geometry is used, and nonlinear absorption coefficient with open aperture. The nonlinear refractive coefficient is calculated from the peak to valley difference of the normalized transmittance by the following formula[5].

$$n_2 = \frac{\Delta \Phi_0}{I_0 L_{eff} K} \tag{3}$$

Where, $K = 2\pi/\lambda$, λ is the beam wavelength, I_o is the intensity at the focal spot, $\Delta\Phi_o$ is the nonlinear phase shift,

$$\Delta T_{p-v} = 0.406 |\Delta \Phi_o| \tag{4}$$

 ΔT_{p-v} the difference between the normalized peak and valley transmittances, L_{eff} is the effective length of the sample, determined from [5]:

$$L_{eff} = \frac{(1 - exp^{-\alpha_0}L)}{\alpha_0} \tag{5}$$

Where L is the sample length, α_0 is given as[6]:

$$\alpha_o = \frac{\ln{(\frac{1}{T})}}{t} \tag{6}$$

Where (t) is the thickness of sample and T is the transmittance. n_o obtained from equation[6]:

$$n_o = \frac{1}{T} + \left[\left(\frac{1}{T^2} - 1 \right) \right]^{1/2} \tag{7}$$

The intensity at the focal spot is given by [6]:

$$I_o = \frac{2P_{peak}}{\pi\omega_o^2} \tag{8}$$

Is defined as the peak intensity within the sample at the focus, where ω_0 is the beam radius at the focal point, the coefficients of nonlinear absorption (β) , can be easily calculated by using following equation[11]:

$$\beta = \frac{2\sqrt{2}\,\mathrm{T}(z)}{\mathrm{I}_0 L_{eff}} \tag{9}$$

Where T(z): The minimum value of normalized transmittance at the focal point, where (z=0).

EXPERIMENTAL WORK

Preparation of Di-Cinnamylidene Benzidine will be explain in detail in the following paragraph:

• Material Preparation

Di-cinnamylidene benzidine was prepared by mixing (1.84 g; 0.01mol) of benzidine dissolved in (10 mL) of absolute ethanol with (2.64 g; 0.02 mol) of cinnamaldehyde dissolved in (10 mL) of absolute ethanol, then three drops of

glacial acetic acid were added to the prepared mixture and left under reflux for (2) hours, producing yellowish solid product.

Figure 1 show preparation of Di-cinnamylidene benzidine.

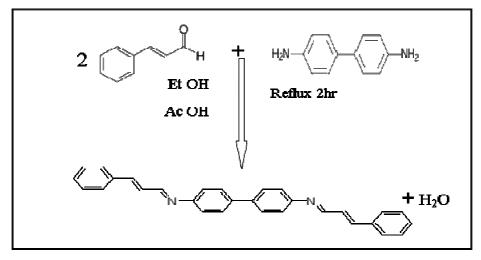


Figure 1: Preparation Design of Di-Cinnamylidene Benzidine

Solutions of concentrations $(1x10^{-3} \text{ M})$ for (Di- Cinnamylidene Benzidine), in ethanol solvent were prepared. The powder was weighed by using an electronic balance type (BL 210 S), Germany, having a sensitivity of four digits. Different concentrations were prepared according to the following equation:

$$W = \frac{M_W \times V \times C}{1000} \tag{10}$$

Where, W: Weight of the dissolved in material (g), Mw: Molecular weight of the material

(g/mol),V: Volume of the solvent (mL), C: The concentration (M).

The prepared solutions were diluted according to the following equation:

$$C_1 V_1 = C_2 V_2 \tag{11}$$

Where: C_1 : Primary concentration, C_2 : New concentration. V_1 : The volume before dilution, V_2 : The volume after dilution. In this work, six concentrations were prepared for (Di-Cinnamylidene Benzidine), the concentrations were $(9\times10^{-5}.8\times10^{-5}.7\times10^{-5}.6\times10^{-5}.5\times10^{-5}$ and 4×10^{-5}) M.

Absorption And Transmission Spectra

The linear absorption and transmission spectra of (Di-Cinnamylidene Benzidine) at different concentrations recorded for wavelengths (190 to 900) nm were tested using UV-VIS spectrophotometer model (Aquarius 7000, Optima, Japan), at room temperature, as shown in Figures 2 and 3. The absorption spectra show two clear bands, the first at wavelength (290) nm, is due to the phenomena of positional excitation for the benzene rings because the absorption happened for the (C=C) group of the aromatic system and azomethane (C=N) group, that resulted from an electronic transitions for $(\pi \rightarrow \pi^*)$. The electronic transitions between azomethane groups and aromatic rings (Phenyl-N) and (Phenyl-C).

The second band between (340-400) nm with highest wavelengths belongs to transition of $(n \rightarrow \pi^*)$ that referred

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to the neighboured active azomethine groups, which acted as an electron-receptor where the free lone pair of electron was available at each nitrogen atom (has ability for sharing at resonance phenomena).

The present results show that the absorption peaks for (Di-Cinnamylidene Benzidine) of different concentrations of in ethanol solvent were shifted toward the longer wavelengths with decreasing concentrations. This shift obtain due to decreasing number of molecules per volume unit at low concentrations, we show absorption increasing with increases concentration. The optical transmission of (Di-Cinnamylidene Benzidine) are shown a variable behavior of the transmission as a function of the incident wavelength.

The linear refractive coefficient (n_o) and linear absorption coefficient (α_o) of (Di-Cinnamylidene Benzidine), obtained from equations (6,7) respectively [9]. The values of (α_o) and (n_o) are decreased with decreasing the concentrations of solutions as listed in Table 1.

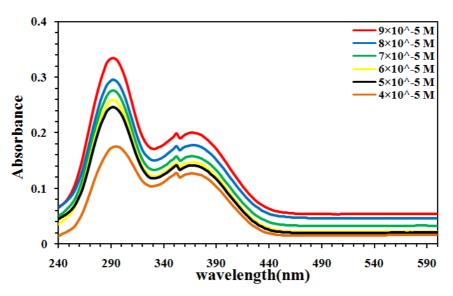


Figure 2: Absorption Spectra of (Di-Cinnamylidene Benzidine) in Ethanol Solvent at Different Concentrations

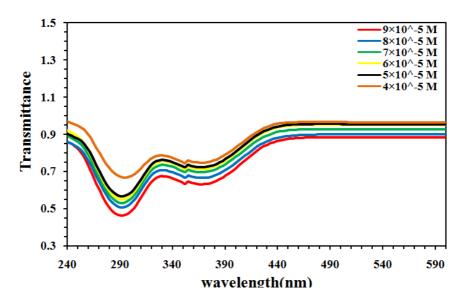


Figure 3: Transmission Spectra of (Di-Cinnamylidene Benzidine) in Ethanol Solvent at different Concentrations

RESULTS AND DISCUSSIONS

The Z-scan experiments will be explain in detail in the following paragraph:

• Z-Scan Measurements

The Z-scan experiments were performed using a continuous wave (CW) diode solid state laser at 473 nm wavelength and 20 mW power, which was focused by (15mm) focal length lens. The schematic of the experimental set up used is shown in Figure 4. A(1mm) wide optical cell containing the solution of (Di-Cinnamylidene Benzidine) is translated across the focal region along the axial direction that is the direction of the propagation laser beam. There are two methods of Z-Scan technique, closed aperture to obtain nonlinear refractive coefficient, and open aperture method to obtain nonlinear absorption coefficient. The far field intensity is measured as a function of the sample position by properly monitoring the transmittance change through a small aperture at the far field position (closed aperture) [7].

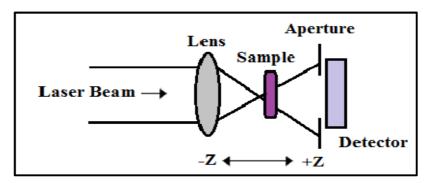


Figure 4: Schematic Diagram of Experimental Arrangement for the Z-Scan Measurement[7]

The third-order nonlinear refractive index n_2 , and the nonlinear absorption coefficient, β , of the (Di-Cinnamylidene Benzidine) in ethanol solvent at different concentrations were evaluated by the measurements of Z-Scan. The intensity at the focal spot is $(I_0 = 20.408 \text{ kW/cm}^2)$.

• Nonlinear Optical Properties

The nonlinear properties were measured by extracting the nonlinear refractive coefficient (n_2) by closed-aperture Z-Scan measurements and nonlinear absorption coefficient (β) by open-aperture Z-Scan. The nonlinear refractive index of the (Di-Cinnamylidene Benzidine) in ethanol solvent at different concentrations (9×10^{-5} , 8×10^{-5} , 7×10^{-5} , 6×10^{-5} , 5×10^{-5} , and 4×10^{-5}) M, were measured by the Z-Scan technique. The measurements were done at 473nm,20 mW. Figure 5 shows closed-aperture Z-Scan at different concentrations of (Di-Cinnamylidene Benzidine) in ethanol solvent at 473nm, 20 mW, the nonlinear effect region is extended from (-5) mm to (5) mm and the transmittance difference between peak and valley, ΔT_{p-v} at concentrations (9×10^{-5} , 8×10^{-5} , 7×10^{-5} , 6×10^{-5} , 5×10^{-5} ,and 4×10^{-5}) M equals to (0.038, 0.032, 0.027, 0.021,0.015,and 0.011) respectively. High transmittance is (0.9657) and low transmittance is (0.8823) at concentration (9×10^{-5}) M.

To investigate the nonlinear absorption coefficient, Figure 6 shows open-aperture Z-Scan for (Di-Cinnamylidene Benzidine) in ethanol solvent at different concentrations at 473nm, 20 mW. Its noticed the valley was smaller and the minimum transmittance T(z) are (0.8620, 0.8470,0.8230,0.8000,0.7800 and 0.7400) at different concentrations (9×10⁻⁵, 8×10⁻⁵,7×10⁻⁵, 6×10⁻⁵, 5×10⁻⁵, and 4×10⁻⁵) M respectively, also from Figure 6 noticed two photon absorption phenomenon. This behaviour is in agreement with [4,6].

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In order to represent the behavior of the material when it is put in the open z-scan system, must follow the following steps. At different distances from the far field of the sample position (-Z), the transmittance is constant with distance. At the near field the transmittance curve begins to decrease until it reaches the minimum value T(z) at the focal point, where (Z=0) mm. The transmittance begins to increase toward the linear behavior at the far field of the sample position (+Z). The change of the intensity in this case is caused by two photon absorption in the sample travels through beam waist.

The nonlinear parameters are calculated, as tabulated in Table 1, from this Table we show that the values of nonlinear parameters (n_2 and β) are decreased with decreasing the concentrations of (Di-Cinnamylidene Benzidine) in ethanol solvent, as decreasing the values of linear parameters (α_0 and α_0). This is due to decreasing number of molecules per volume unit at low concentrations.

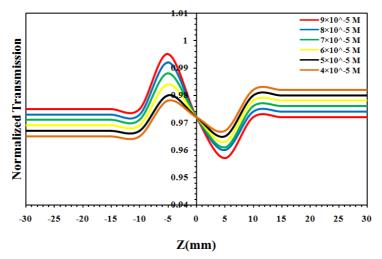


Figure 5: Closed-Aperture Z-Scan Data for (Di-Cinnamylidene Benzidine) in Ethanol Solvent at Different Concentration

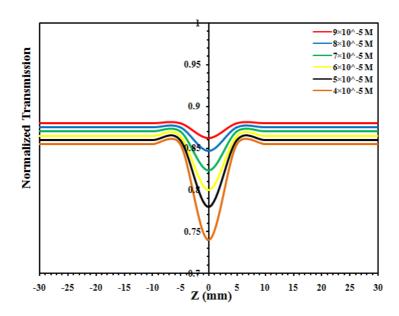


Figure 6: Open-Aperture Z-Scan Data for (Di-Cinnamylidene Benzidine) in Ethanol Solvent at Different Concentration

Concent ration ×10 ⁻⁵ (M)	Linear Transmission T	Linear Absorption Coefficient a ₀ (cm ⁻¹)	Linear Refractive Index n _o	ΔT _{p-v}	$\Delta \phi_{\circ}$	n ₂ ×10 ⁻¹⁰ (cm ² /mW)	T(z)	β ×10 ⁻³ (cm/mW)
9	0.8823	0.1252	1.6668	0.038	0.0935	3.4750	0.8620	1.1946
8	0.8975	0.1081	1.6054	0.032	0.0788	2.7434	0.8470	1.1750
7	0.9270	0.0757	1.4829	0.027	0.0666	2.4666	0.8230	1.1372
6	0.9501	0.0511	1.3808	0.021	0.0517	1.8937	0.8000	1.0912
5	0.9545	0.0464	1.3597	0.015	0.0369	1.3566	0.7800	1.0682
4	0.9657	0.0348	1.3039	0.011	0.0270	1.0030	0.7400	1.0204

Table 1: The Nonlinear Optical Parameters for (Di-Cinnamylidene Benzidine) in Ethanol Solvent at Different Concentrations

Optical Limiting Behavior

Optical limiting occurs when the optical transmission of a material saturates with increasing laser intensity, a property that is desirable for the protection of sensors and human eyes from the intense laser radiation. The optical power limiting property of the (Di-Cinnamylidene Benzidine) in ethanol solvent at different concentrations, is measured with the same laser used in Z-Scan technique. Additionally a varying beam splitter was used to vary the input power. Figure 7 gives the optical limiting characteristics at room temperature for the sample. The sample shows very good optical limiting behavior arising from nonlinear refraction, the sample starts defocusing the beam resulting in a greater part of the beam cross-section being cut off by the aperture. Thus the transmittance recorded by the photo detector remained reasonably constant showing a plateau region. This indicates that the properties of optical limiting become better with increasing the concentrations. This behavior is in agreement with [4].

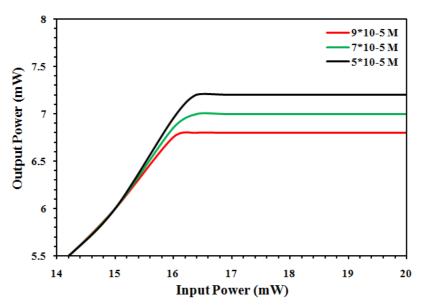


Figure 7: Optical Limiting Response (Di-Cinnamylidene Benzidine) in Ethanol Solvent at Different Concentrations at 473 Nm

CONCLUSIONS

We have measured the nonlinear refraction index n_2 and the nonlinear absorption coefficient β for the solutions of (Di-Cinnamylidene Benzidine) in ethanol solvent for different concentrations, using the Z-Scan technique with 473 nm of

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(CW) laser. The Z-Scan measurements indicated that the material exhibited large nonlinear optical properties. We have shown that the nonlinear absorption can be attributed to a two photon absorption process, while the nonlinear refraction leads to self-defocusing in this material. All the solutions samples showed a large nonlinear refractive coefficient and nonlinear absorption coefficient of the order of 10^{-10} cm²/mW and 10^{-3} cm/mW, respectively, the optical limiting behavior has been studied. All these experimental results show that the solution (Di-Cinnamylidene Benzidine) is a promising material for applications in nonlinear optical devices.

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